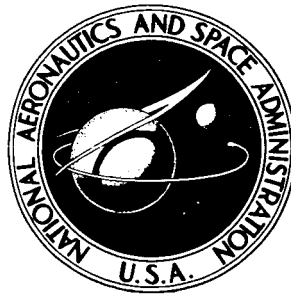


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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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# INFLUENCE OF PARENT AND DAUGHTER DECAY ON TRACER DIFFUSION PROFILES

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## SUMMARY

In some radioactive tracer diffusion experiments, the diffusion profile is determined by sectioning the sample and counting the gamma activity of each of the sections. If the daughter decay product is radioactive and has in its decay chain a gamma ray whose energy is close to that of the tracer, a correction to the counting rates of the sections is necessary. Because the daughter decay product is a chemical species different from the tracer, its diffusion coefficient will, in general, be different from the tracer. An equation is developed which gives the diffusion profile for the daughter for a one-dimensional diffusion problem. This solution can be used in making an estimate of the error produced by the daughter decay. With sufficient knowledge of the diffusing species, the correction to the diffusion profile can be made.

## INTRODUCTION

A method commonly used to determine the diffusion profile of a radioactive tracer in a one dimensional diffusion experiment is to section the sample and to measure the activity of each of the sections by observing the decay rate of tracer to its daughter. If the daughter is radioactive and its activity is included with that of the parent, the resulting diffusion profile will be incorrect. This problem has not been encountered very frequently in the past because it has been possible to choose tracers which have stable decay products, or, if the decay products are radioactive, to exclude by some means their influence on the counting rate due to the tracer. More recently, work on isotope effects has been done in which the dependence of diffusion on the mass of the diffusing atom is studied. This requires pairs of radioactive tracers of the same chemical species which can be diffused simultaneously into a specimen and distinguished by the characteristics of their decay. This more stringent requirement may necessitate the use of tracers that decay into radioactive daughters.

An example where this occurs is the self diffusion of zirconium using zirconium 95

as a tracer. The decay product of zirconium 95 is niobium 95 which emits a gamma ray whose energy is close to that of zirconium 95.

## CALCULATION

Let  $n_p(x, t)$  and  $n_d(x, t)$  be, respectively, the volume concentrations of the parent (the tracer) and the daughter decay product as a function of the distance  $x$  and time  $t$  where  $x$  is measured from the surface of the semi-infinite sample. One dimensional diffusion is considered so that the concentrations  $n_p(x, t)$  and  $n_d(x, t)$  do not depend on  $y$  and  $z$ . Let the parent have a decay constant  $\lambda_p$  and a diffusion coefficient  $D_p$ ; similarly, the daughter has a decay constant  $\lambda_d$  and a diffusion coefficient  $D_d$ . The concentration  $n_p(x, t)$  of the parent then satisfies the differential equation

$$\frac{\partial n_p(x, t)}{\partial t} = D_p \frac{\partial^2 n_p(x, t)}{\partial x^2} - \lambda_p n_p(x, t) \quad (1)$$

This is the usual one dimensional diffusion equation with the addition of the last term which takes into account the rate of decay of the parent. If the parent is considered to have an initial concentration  $n_{p0}$  in a narrow region of thickness  $a$  at the surface of the sample, the boundary conditions are

$$\left. \begin{array}{ll} n_p(x, 0) = n_{p0} & 0 \leq x \leq a \\ n_p(x, 0) = 0 & a < x \end{array} \right\} \quad (2a)$$

$$\frac{\partial n_p(0, t)}{\partial x} = 0 \quad (2b)$$

and

$$\lim_{x \rightarrow \infty} n_p(x, t) = 0 \quad (2c)$$

From equation (2a) initially, the amount of the parent per unit area of the sample surface is

$$N_p = n_{p0}a \quad (3)$$

where  $N_p$  has the dimensions of atoms per unit area. If the diffusion distance is sufficiently large compared with the thickness  $a$  of the plating of parent atoms, an approximation is justified in which  $a$  is allowed to approach zero in such a manner that  $N_p$  remains constant. With this approximation, the solution to equation (1) is obtained in the appendix as

$$n_p(x, t) = \frac{N_p \exp\left(-\lambda_p t - \frac{x^2}{4D_p t}\right)}{(\pi D_p t)^{1/2}} \quad (4)$$

Now, calculating the concentration of the daughter, the differential equation

$$\frac{\partial n_d(x, t)}{\partial t} = D_d \frac{\partial^2 n_d(x, t)}{\partial x^2} + \lambda_p n_p(x, t) - \lambda_d n_d(x, t) \quad (5)$$

applies, where  $\lambda_p n_p(x, t)$  is the rate of production of the daughter from the decay of the parent, and  $\lambda_d n_d(x, t)$  is the rate of loss of the daughter due to radioactive decay.

The boundary conditions are

$$\left. \begin{aligned} n_d(x, 0) &= n_{d0} & 0 \leq x \leq a \\ n_d(x, 0) &= 0 & a < x \end{aligned} \right\} \quad (6a)$$

$$\frac{\partial n_d(0, t)}{\partial x} = 0 \quad (6b)$$

and

$$\lim_{x \rightarrow \infty} n_d(x, t) = 0 \quad (6c)$$

where  $n_{d0}$  is the initial concentration of the daughter in the surface plating. Letting  $N_d$  be the initial amount of the daughter per unit area of the sample surface, and, as before, letting  $a \rightarrow 0$  and holding  $N_d$  constant where

$$N_d = n_{d0} a \quad (7)$$

yields the solution to equation (5) (see appendix) as

$$\begin{aligned}
n_d(x, t) = & \frac{N_d \left( \exp -\lambda_d t - \frac{x^2}{4D_d t} \right)}{(\pi D_d t)^{1/2}} \\
& + \frac{\lambda_p N_p \exp - \left( \frac{\lambda_p D_d - \lambda_d D_p}{D_d - D_p} t \right)}{D_d - D_p} \int_0^t \left( \frac{D_d}{\pi \tau} \right)^{1/2} \exp \left[ \frac{D_d (\lambda_p - \lambda_d) \tau}{D_d - D_p} - \frac{x^2}{4D_d \tau} \right] \\
& - \left( \frac{D_p}{\pi \tau} \right)^{1/2} \exp \left[ \frac{D_p (\lambda_p - \lambda_d) \tau}{D_d - D_p} - \frac{x^2}{4D_p \tau} \right] d\tau
\end{aligned} \tag{8}$$

The first term of equation (8) gives the contribution due to the initial surface concentration  $N_d$  of the daughter, and the last term gives the contribution due to the decay of the parent to its daughter during the diffusion anneal itself.

The total counting rate including both parent and daughter is now calculated. The counting rate  $r_{p,d}$  of a thin section of thickness  $\Delta l$  and area  $A$  where the center of the section is a distance  $x$  from the surface is

$$r_{p,d} = \eta A \Delta l \left[ f_p \lambda_p n_p(x, t) + f_d \lambda_d n_d(x, t) \right] \tag{9}$$

where it is assumed that the parent and daughter gamma rays cannot be distinguished. In equation (9),  $\eta$  represents the efficiency of counter for counting the decays and  $f_p$  and  $f_d$  are the fractions of decays of the parent and daughter, respectively, which result in the interfering gamma rays.

## RESULTS

Using a digital computer and numerical integration, the integral in equation (8) was evaluated for various values of the  $D_d$  and using equation (9), the total counting rate was evaluated. The decay constants,  $\lambda_p$  and  $\lambda_d$ , of both parent and daughter were set equal to  $1 \times 10^{-7}$  reciprocal second, a half life of about 80 days, and the diffusion time  $t$  was set equal to  $2 \times 10^6$  second, about 23 days. This anneal time was chosen to be comparable with the isotope half life so that the influence of the daughter on the parent diffusion profile could be clearly shown. The diffusion coefficient for the parent was

$D_p = 1 \times 10^{-9}$  centimeter squared per second, and, because an examination of cases where the diffusion coefficient of the daughter was both larger and smaller than that of the parent was desired, several values of  $D_d$  between  $10^{-6}$  centimeter squared per second to  $10^{-12}$  centimeter squared per second were used. Figure 1 which is a plot of the log of  $r_{p,d}/A \Delta l \eta$  as a function of  $x^2/t$ , shows these results. The short-dashed curves in these figures represent the usual diffusion profile obtained from the counting rate of the parent only. Each of the other curves then represents the diffusion profile obtained by summing the counting rates of the parent and daughter. Figure 1(b), where  $D_p > D_d$ , shows clearly that the slopes derived from the solid curves and hence the apparent diffusion coefficients are only a few percent smaller than the true value given by the reciprocal slope of the dashed line representing the parent alone. However, the influence of the daughter is much more marked in figure 1(b) where  $D_d > D_p$ . In the worst case presented, a diffusion coefficient calculated from a straight line drawn through curves with  $D_d/D_p$  of 10 or 100 could overestimate  $D_p$  by 25 percent. An error in a diffusion coefficient of 5 to 10 percent is of little significance in itself. However, errors arising from neglect of the daughter increase as the temperature of the anneal decreases due to the longer annealing time required to give an adequate diffusion zone. Since the errors are systematic and become larger with decreasing temperature, serious error could result in the measured activation energy for diffusion.

The problem produced by the activity due to daughter decay becomes most serious in mass dependence studies where the critical measurement is the change in the relative concentration of two diffusing isotopes as a function of the depth of penetration. Here, one is attempting to measure changes in relative concentration of only a few percent. Ignoring the contribution of even a small quantity of daughter activity in one of the tracer isotopes might easily lead to such large errors in the mass effect that the results of the experiment would be meaningless.

Lewis Research Center,  
 National Aeronautics and Space Administration,  
 Cleveland, Ohio, May 15, 1967,  
 129-03-15-01-22.

## APPENDIX - SOLUTION OF DIFFERENTIAL EQUATION

### Parent Case

In obtaining the solution to equations (1) and (5) we have used the method of Laplace transforms. Equation (1) along with the boundary conditions becomes

$$\frac{\partial n_p(x, t)}{\partial t} = D_p \frac{\partial^2 n_p(x, t)}{\partial x^2} - \lambda_p n_p(x, t)$$

$$n_p(x, 0) = n_{p0} [1 - S_a(x)]$$

and

$$\frac{\partial n_p(0, t)}{\partial x} = 0 \tag{A1}$$

$$\lim_{x \rightarrow \infty} n_p(x, t) = 0$$

where the function  $S_a(x)$  is defined as

$$\begin{aligned} S_a(x) &= 0 & 0 \leq x \leq a \\ &= 1 & a < x \end{aligned} \tag{A2}$$

Transforming equation (A1) on  $t$  and letting  $f(x, s)$  be the Laplace transform of  $n_p(x, t)$  yield

$$sf(x, s) - n_{p0} [1 - S_a(x)] = D_p \frac{\partial^2 f(x, s)}{\partial x^2} - \lambda_p f(x, s) \tag{A3}$$

where the boundary condition at  $t = 0$  was used. Letting  $g(r, s)$  be the Laplace transform of  $f(x, s)$  on  $x$ , equation (A3) transforms into



$$sg(r, s) - \frac{n_{p0}}{r} [1 - \exp(-ar)] = D_p [r^2 g(r, s) - rf(0, s)] - \lambda_p g(r, s) \quad (A4)$$

where the boundary condition at  $x = 0$  was used.

Equation (A4) can then be solved for  $g(r_{p,d}, s)$  and using standard methods the inverse Laplace transform on  $r_{p,d}$  is obtained as

$$f(x, s) = \frac{N_p \exp \left[ - \left( \frac{s + \lambda_p}{D_p} \right)^{1/2} x \right]}{[D_p (s + \lambda_p)]^{1/2}} \quad (A5)$$

where  $a \rightarrow 0$  such that

$$an_{p0} = N_p \quad (A6)$$

and  $N_p$  is the surface concentration of the parent. Also, in obtaining equation (A5),  $f(0, s)$  has been eliminated by using the boundary condition for  $x \rightarrow \infty$ . Obtaining the inverse transform of  $f(x, s)$  on  $s$  in equation (A5) yields

$$n_p(x, t) = \frac{N_p \exp \left( - \lambda_p t - \frac{x^2}{4D_p t} \right)}{(\pi D_p t)^{1/2}} \quad (A7)$$

The effect of the parent decay on the concentration is to multiply the usual result by a factor  $\exp(-\lambda_p t)$ .

## DAUGHTER CASE

The differential equation for the concentration of the daughter is from equation (5).

$$\frac{\partial n_d(x, t)}{\partial t} = D_d \frac{\partial^2 n_d(x, t)}{\partial x^2} + \lambda_p n_p(x, t) - \lambda_d n_d(x, t)$$

$$n_d(x, 0) = n_{d0}[1 - S_a(x)]$$

$$\frac{\partial n_d(0, t)}{\partial x} = 0$$

and

$$\lim_{x \rightarrow \infty} n_d(x, t) = 0 \quad (A8)$$

The Laplace transform on  $t$  of (A8), where  $f(x, s)$  is the transform of  $n_d(x, t)$ , is

$$sf(x, s) - n_{d0}[1 - S_a(x)] = D_d \frac{\partial^2 f(x, s)}{\partial x^2} + \frac{\lambda_p N_p \exp - \left( \frac{s + \lambda_p}{D_p} \right)^{1/2} x}{[D_p(s + \lambda_p)]^{1/2}} - \lambda_d f(x, s) \quad (A9)$$

where the boundary condition at  $t = 0$  has been used. Defining  $g(r, s)$  as the Laplace transform of  $f(x, s)$  on  $s$  yields for equation (A9)

$$\begin{aligned} sg(r, s) - \frac{n_{d0}}{r} [1 - \exp(-ar)] &= D_d [r^2 g(r, s) - rf(0, s)] \\ &+ \frac{\lambda_p N_p}{[D_p(s + \lambda_p)]^{1/2} \left( r + \frac{s + \lambda_p}{D_p} \right)^{1/2}} - \lambda_d g(r, s) \end{aligned} \quad (A10)$$

where the boundary condition at  $x = 0$  was used. By letting  $a \rightarrow 0$  in equation (A10) in such a manner that

$$an_{d0} = N_d \quad (A11)$$

where  $N_d$  is the surface concentration of the daughter at  $t = 0$ , equation (A10) can be solved for  $g(r, s)$ . Taking the inverse transform on  $r$  and eliminating  $f(0, s)$  by using the boundary condition for  $x \rightarrow \infty$  yield

$$\begin{aligned}
f(x, s) = & \frac{N_d \exp \left[ - \left( \frac{s + \lambda_d}{D_d} \right)^{1/2} x \right]}{[D_d(s + \lambda_d)]^{1/2}} \\
& + \frac{\lambda_p N_p}{D_p D_d} \left\{ \frac{\exp \left[ - \left( \frac{s + \lambda_d}{D_d} \right)^{1/2} x \right]}{\left( \frac{s + \lambda_d}{D_d} \right)^{1/2} \left( \frac{s + \lambda_p}{D_p} - \frac{s + \lambda_d}{D_d} \right)} - \frac{\exp \left[ - \left( \frac{s + \lambda_p}{D_p} \right)^{1/2} x \right]}{\left( \frac{s + \lambda_p}{D_p} \right)^{1/2} \left( \frac{s + \lambda_p}{D_p} - \frac{s + \lambda_d}{D_d} \right)} \right\}
\end{aligned} \tag{A12}$$

Obtaining the inverse transform on  $s$ , yields

$$\begin{aligned}
n_d(x, t) = & \frac{N_d \exp \left[ - \lambda_d t - \frac{x^2}{4D_d t} \right]}{(\pi D_d t)^{1/2}} \\
& + \frac{\lambda_p N_p \exp \left( - \frac{\lambda_p D_d - \lambda_d D_p}{D_d - D_p} \right)}{D_d - D_p} \int_0^t \left\{ \left( \frac{D_d}{\pi \tau} \right)^{1/2} \exp \left[ \frac{D_d (\lambda_p - \lambda_d) \tau}{D_d - D_p} - \frac{x^2}{4D_d \tau} \right] \right. \\
& \left. - \left( \frac{D_p}{\pi \tau} \right)^{1/2} \exp \left[ \frac{D_p (\lambda_p - \lambda_d) \tau}{D_d - D_p} - \frac{x^2}{4D_p \tau} \right] \right\} d\tau
\end{aligned} \tag{A13}$$

